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The Patent Office

Cardiff Road
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1. Your reference — 15438 LgCm

2. Patent application number *(The Patent Office will fill in this part)* — **9924999.7** — 22 OCT 1999

3. Full name, address and postcode of the or of each applicant *(underline all surnames)*

AEA Technology plc
329 Harwell
Didcot, Oxfordshire, OX11 0RA
United Kingdom

Patents ADP number *(if you know it)* 6969372001

If the applicant is a corporate body, give the country/state of its incorporation — England and Wales

4. Title of the invention

Reactor for the plasma treatment of gases

5. Name of your agent *(if you have one)*

Marcus John Lofting

"Address for service" in the United Kingdom to which all correspondence should be sent *(including the postcode)*

AEA Technology plc
Patents Department, 329 Harwell
Didcot,Oxfordshire,OX11 0RA

Patents ADP number *(if you know it)* 7298474001

6. If you are declaring priority from one or more earlier patent applications, give the country and the date of filing of the or of each of these earlier applications and *(if you know it)* the or each application number

Country	Priority application number *(if you know it)*	Date of filing *(day / month / year)*

7. If this application is divided or otherwise derived from an earlier UK application, give the number and the filing date of the earlier application

Number of earlier application	Date of filing *(day / month / year)*

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b) there is an inventor who is not named as an applicant, or
c) any named applicant is a corporate body.
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Yes

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Continuation sheets of this form	
Description	6
Claim(s)	3 DML
Abstract	1
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Statement of inventorship and right to grant of a patent *(Patents Form 7/77)*	
Request for preliminary examination and search *(Patents Form 9/77)*	
Request for substantive examination *(Patents Form 10/77)*	
Any other documents *(please specify)*	

11. I/We request the grant of a patent on the basis of this application.

Signature M. Lofting Date 22.10.99

M.J. LOFTING (On behalf of AEA Technology plc by virtue of a Power of Attorney dated 26th March 1996)

12. Name and daytime telephone number of person to contact in the United Kingdom 01235 432037 Mrs C A Cassidy

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Abstract

Reactor for Plasma Treatment of Gases

In a non-thermal plasma reactor, at least a component of the active material is selected or modified to provide the capability to adsorb or trap a predetermined chemical species in the gas flow thereby to increase the effective residence time of said species relative to the residence time of unadsorbed species in the gas flow.

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Reactor for Plasma Treatment of Gases

The invention relates to a reactor for plasma treatment of gases and more particularly to a method of manufacturing a reactor utilising a non-thermal plasma.

There is increasing interest in the use of non-thermal plasmas for treatment of gaseous exhausts and in particular for treatment of exhausts from motor vehicles. Treatment of exhausts involves conversion of harmful exhaust components such as hydrocarbons to carbon dioxide and water as well as harmful NO_x components of exhausts to nitrogen. In the case of lean burn engines such as diesels there is a requirement for removal of carbonaceous particulates by for example oxidation to carbon dioxide. Examples of non-thermal treatment of exhausts are described in US 3,983,021 (Monsanto), US 5,147,516 (Tecogen) and US 5,254,231 (Battelle Memorial Institute). GB 2,274,412 (AEA Technology) describes a method for the treatment of diesel emissions by a non-thermal plasma for oxidation of carbonaceous particulates and reduction of NO_x to nitrogen.

Plasma can be used to activate or produce reactant species, which then subsequently react with or without catalytic enhancement to yield the desirable products. For example, our publication WO99/12638 describes the plasma production of plasma activated hydrocarbons as a precursor to the selective catalytic reduction of NO_x to N_2. Examples of catalysts for this selective reduction of NO_x to nitrogen are ceramics such as aluminas, zirconias, titanias, zeolites, metal-exchanged zeolites particularly those with a hydrothermal stability characterised by an absence of Bronsted acid groups,

precious metal doped oxides such as aluminas as described in WO99/12638. Examples of suitable catalysts are also described in an article 'Selective catalytic reduction of NOx with N-free reductants' by M Shelef published in Chem Rev 1995, pp209-225.

In such catalytically enhanced plasma processes the reactant species such as plasma activated hydrocarbons, which can include oxygenated hydrocarbons, are often produced as intermediates in e.g. the stepwise decomposition of hydrocarbons by reactions of O_2, O, OH and HO_2. Where the reactants are intermediates and the catalyst is highly selective to a given reductant, optimisation of the process can become difficult, if not impossible. This is because the completeness of the plasma reactions and hence the concentration of intermediates is controlled largely by the input power (joules per second) for a given residence time in the plasma reactor. This leads to the idea of a normalised unit expressed in joules per litre, which will determine the concentration of intermediates in a plasma reactor. For example in an article 'Plasma assisted catalytic reduction of NO_x by BM Penetrante et al, SAE 982508, it is shown how the gas phase composition changes with input power expressed as joules per litre of gas volume for an exhaust flow in litres per unit time. This key parameter, joules/litre, is largely fixed by vehicle constraints, i.e. reactor size, and the acceptable level of power input to the reactor in as far as all of the exhaust passes through the reactor.

The present invention is based upon an appreciation of the advantages that follow if one changes the residence time of selected species in the reactor, and thus breaks the, at present unavoidable, link between

joules per litre input power and reactant species. This would lead to a simplification in the design of plasma reactors.

Examples have been given which show that, without catalytic enhancement, plasma reactors can produce quantities of undesirable by-products usually associated with partial oxidation of hydrocarbons (see 'Analysis of plasma-catalysis for diesel NO_x remediation' by J Hoard and ML Balmer, SAE 982429), for example methyl nitrate, formaldehyde. A solution is offered if hydrocarbons can be retained for relatively long periods of time to achieve complete conversion to CO and CO_2, while oxides of nitrogen may require a short residence time to avoid formation of acids.

It is an object of the present invention to provide a method of manufacturing a component for a non-thermal plasma reactor which addresses these problems.

The invention provides, in one of its aspects, a method of manufacturing a component for a non-thermal plasma reactor for the treatment of gases, which method comprises assembling a bed of active material in an enclosure having gas flow conduits for directing gas to flow through or over the bed of active material, providing electrodes adapted when electrically energised to generate non-thermal plasma in the gas, at least a component of the active material being selected for its capability to adsorb or trap a predetermined chemical species in the gas flow thereby to increase the effective residence time of said species relative to the residence time of species in the gas flow which are not adsorbed or trapped.

The invention includes a non-thermal plasma reactor for the treatment of gases, comprising a bed of active material in an enclosure having gas flow conduits for directing gas to flow through or over the bed of active material, electrodes adapted when electrically energised to generate non-thermal plasma in the gas, at least a component of the active material being modified so as to adsorb or trap a predetermined chemical species in the gas flow to increase the effective residence time of the said species relative to the residence time of species in the gas flow which are not adsorbed or trapped.

The function of the active material, or component thereof, having the capability to adsorb selected species can be seen as that of a selective filter for that species.

In addition to the advantages, referred to above, which result from the effective increase in residence time of the selected species, such a selective filter can operate to trap the reactants and hold them for sufficient time for them to be activated by a plasma to a state where they can react with NO_x to yield desirable products. In this role the filter material in the presence of a plasma can be made to appear to act as a catalytic surface but importantly neither the plasma nor the selective filter alone need have catalytic properties.

A selective filter according to the present invention provides, by selective modification of residence times, a method of controlling and hence optimising the distribution of product species from a plasma reactor with a significant degree of independence from flow rate, reactor size or energy density.

In a specific embodiment of the invention described by way of example, the structure of the reactor and its bed of active material is as described in our patent specification WO99/12638, or the specification of our application GB 99 11728.5. The active material comprises polymeric or ceramic material (dielectric or ferroelectric) in the form of sheets, wafers, meshes, frits, coils, spheres, pellets, extrudates, granules, fibres, foams or honeycomb monolith or as a coating on sheets, wafers, meshes, frits, coils, spheres, pellets, extrudates, granules, fibres or honeycomb monolith, foam, honeycomb monolith or membrane in the plasma region of the non-thermal plasma reactor. At least a component of the active material is selected or modified in order to adsorb or trap a predetermined chemical species in the gas flow thereby to increase the effective residence time in the reactor of the said species relative to the residence time of unadsorbed species in the gas flow. The active material may comprise a plurality of components each of which adsorbs or traps a different chemical species thereby to increase the effective residence time in the reactor of each of the different adsorbed chemical species. It will be appreciated that a selected or modified component of the active material may adsorb or trap more than one selected chemical species. Also, where a plurality of components is provided each of which adsorbs or traps a different chemical species, the respective adsorptions may be different for each different adsorbed species, thus providing a correspondingly different increase in the effective residence time of each of the different adsorbed species.

Suitable non-thermal plasma reactors are those of the ferroelectric bed type, dielectric barrier discharge type, pulsed corona discharge reactor or surface discharge reactor.

When a gas molecule, for example, hydrocarbon enters a non-thermal plasma it resides in the plasma zone for a given period of time known as the residence time whereupon chemical reactions can occur. When a selective filter in accordance with the present invention is present in the plasma region the residence time of gas molecules in the plasma region increases. This is because a gas molecule on entering the plasma zone can adsorb onto the surface of material in the bed and desorb at a later time. The molecule now in the gas phase can readsorb onto different regions of the bed. This process of adsorption, desorption and readsorption can occur throughout the plasma region thus increasing residence times of molecules entering the plasma region allowing chemical reactions to occur more extensively. In another example the selective filter may adsorb species prior to plasma activation and/or surface decomposition and subsequent desorption. The material composition, e.g. acidity (Bronsted and Lewis acid sites), pore size, pore shape, pore size distribution, surface area are characteristics which can be modified to effect different residence times and selectivity.

Claims

1. A method of manufacturing a component for a non-thermal plasma reactor for the treatment of gases, which method comprises assembling a bed of active material in an enclosure having gas flow conduits for directing gas to flow through or over the bed of active material, providing electrodes adapted when electrically energised to generate non-thermal plasma in the gas , at least a component of the active material being selected for its capability to adsorb or trap a predetermined chemical species in the gas flow thereby to increase the effective residence time of said species relative to the residence time of species in the gas flow which are not adsorbed or trapped.

2. A method as claimed in claim 1, wherein the bed of active material is provided in the form of sheets, wafers, meshes, frits, coils, spheres, pellets, extrudate, granules, fibres, foams or honeycomb monolith or as a coating on sheets, wafers, meshes, frits, coils, spheres, pellets, extrudates, granules, fibres or honeycomb monolith, foam, or membrane.

3. A method as claimed in claim 2, wherein the active material comprises dielectric or ferroelectric material.

4. A method as claimed in claim 2 or claim 3, wherein the active material comprises polymeric material.

5. A method as claimed in claim 2 or claim 3, wherein the active material comprises ceramic material.

6. A method as claimed in any of the preceding claims, wherein dielectric barrier material between the electrodes establishes a dielectric barrier discharge type of reactor.

7. A method as claimed in any of claims 1 to 5, wherein the electrodes are configured to provide a pulsed corona discharge type of reactor.

8. A method as claimed in any of claims 1 to 5, wherein the electrodes are configured to provide a surface discharge type of reactor.

9. A method as claimed in any of the preceding claims, wherein the electrodes are configured to generate plasma in the gas as it flows over or through the bed of active material.

10. A non-thermal plasma reactor for the treatment of gases when made by a method as claimed in any of the preceding claims.

11. A non-thermal plasma reactor for the treatment of gases, comprising a bed of active material in an enclosure having gas flow conduits for directing gas to flow through or over the bed of active material, electrodes adapted when electrically energised to generate non-thermal plasma in the gas, at least a component of the active material being modified so as to adsorb or trap a predetermined chemical species in the gas flow to increase the effective residence time of the said species relative to the residence time of species in the gas flow which are not adsorbed or trapped.

12. A non-thermal plasma reactor as claimed in claim 11, wherein neither the plasma nor the said modified component is separately catalytic with respect to reactions of chemical species in the gas, but by virtue of the combined effect of the plasma and the adsorption or entrapment a catalytic effect on reactions is achieved.

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